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Experiment Report Form

The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF. This double-page report will be reduced by ESRF to a one page, A4 format, and will be published in the Annex to the ESRF Annual Report.

Should you wish to make more general comments on the experiment, enclose these on a separate sheet, and send both the Report and comments to the User Office.

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- bear in mind that the report will be reduced to 71% of its original size. A type-face such as “Times”, 14 points, with a 1.5 line spacing between lines for the text produces a report which can be read easily.

Note that requests for further beam time must always be accompanied by a report on previous measurements.

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ESRF	**Experiment title:** EXAFS characterization of Ga and Fe extra-framwork species in zeolites	**Experiment number**: 01-01-255
Beamline: BM01B	**Date of experiment**: from: 01 June 2001 to: 04 June 2001	**Date of report**: 27/09/2001
Shifts: 9	**Local contact(s)**: Mr Hermann EMERICH	*Received at ESRF:*

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Report:

Introduction

Emissions of the greenhouse gas N_2O from nitric and adipic acid plants pose a serious environmental problem. Catalytic decomposition of N_2O with Fe-ZSM-5 catalysts seems a very promising option to abate these emission. It has been reported that, in contrast to other materials, Fe-ZSM-5 maintains a high activity for N_2O decomposition even in the presence of NO, O_2 and water, which are all components of a real tail gas [1]. We focused our EXAFS investigation on the interaction of N_2O with the catalyst to understand the reaction mechanism of the conversion of N_2O to N_2.

Experimental

Fe-loaded ZSM-5 zeolites were prepared by sublimation of $FeCl_3$, followed by washing and calcination in O_2. The sample was pressed into a self-supported pellet, placed in the EXAFS and subjected to a thermal treatment at 673 K in pure He or a mixture of 1000 ppm N_2O in He. After cooling down, the Fe K edge spectra were measured in transmission mode at liquid nitrogen temperature. The data was analyzed by standard procedures using the EXAFS Data Analysis Program XDAP- Version 2.2.3.

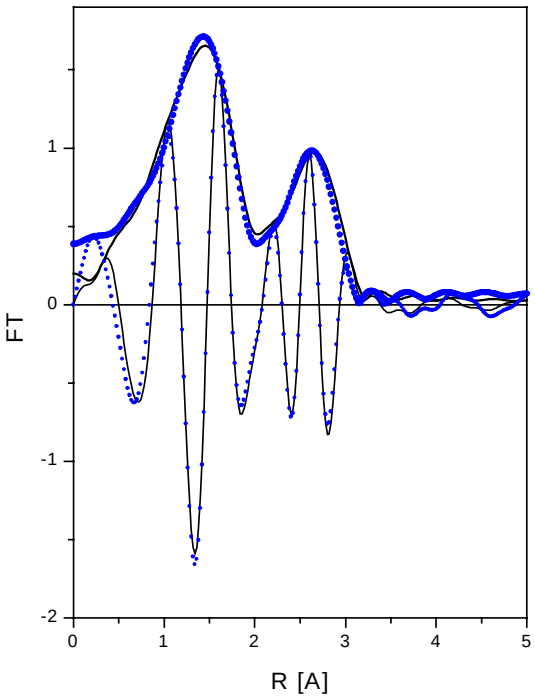

Fig. 1

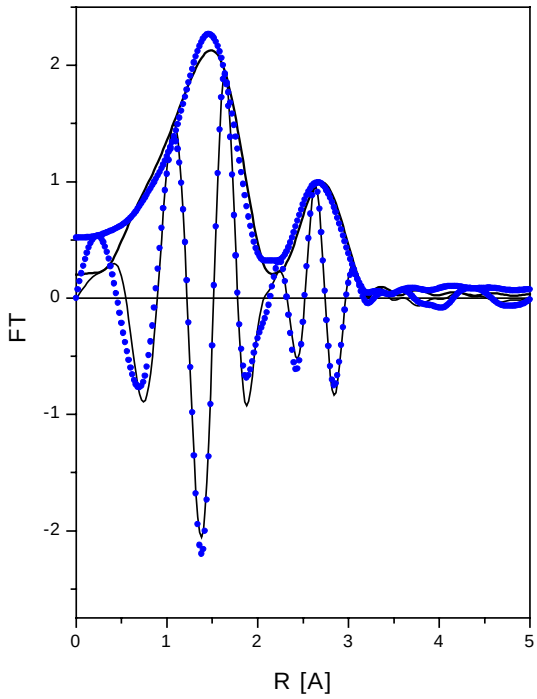

Fig. 2

Results and Discussion

The fourier transformed spectrum (experimental and fitted) of the sample recorded after thermal treatment in He flow at 673 K is shown in Fig.1. The data analysis of the first coordination shell, assigned to the Fe-O backscattering pair, showed that four oxygen atoms are coordinated to Fe atom at a distance of 1.92 Å. The second, Fe-Fe shell, was found at a distance of 2.97 Å. The coordination number of 1.5 revealed the presence of very small iron clusters.

The spectrum detected after the exposure of the catalyst to N_2O is presented in Fig.2. The Fe-O and Fe-Fe shells are determined in the spectrum. The coordination number for the first shell increased and 4.8 oxygen atoms are coordinated to the iron atom at a distance of 1.95 Å. A relatively high Debye –Waller factor obtained for this shell points to the fact that more different distances are included in this shell . This shell will be therefore further investigated to improve the quality of the fit. The iron shell at a distance of 3.01 Å and with the coordination number of 1.7 shows that small Fe clusters are still present after the exposure to N_2O.

The tentative interpretation of the results is that, in the first step of N_2O decomposition, the O from N_2O is added to the coordination shell of the Fe site, i.e.

$FeO_x + N_2O \rightarrow FeO_{x+1} + N_2$

Further measurements have to be done to conclude whether these results support the formation of a peroxide species, as proposed by Sachtler et al. [2].

References

[1] J. Perez-Ramirez, F. Kapteijn, G. Mul, J.A. Moulijn, Chem. Comm. 2001, 693.

[2] Z.-X. Gao, H.-S. Kim, Q. Sun, P. C. Stair, W.M.H. Sachtler, J. Phys. Chem. B 2001, 105, 6186.